

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In re Application of:

Denis UZIO et al.

Examiner: John Christopher Douglas

Serial No.: 10/765,840

Group Art Unit: 1764

Filed: January 29, 2004

Title: PARTIALLY COKED CATALYSTS THAT CAN BE USED IN THE  
HYDROTREATMENT OF FRACTIONS THAT CONTAIN SULFUR-  
CONTAINING COMPOUNDS AND OLEFINS

**APPEAL BRIEF**

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Sir:

This Brief on Appeal is in response to the decision of the Examiner finally rejecting claims 1-14 of the above-identified application. The requisite fee for filing this Brief on Appeal of \$500.00 is attended to by credit card.

**(i) REAL PARTY IN INTEREST**

The present application is assigned to Institut Francais du Petrole by an assignment recorded in the United States Patent and Trademark Office on January 29, 2004, at reel 014939, frame 0299.

**(ii) RELATED APPEALS AND INTERFERENCES**

There are no related appeals or interferences.

**(iii) STATUS OF CLAIMS**

Claims 1, and 3-14 are pending and are all on appeal. Claim 2 has been cancelled.

**(iv) STATUS OF AMENDMENTS**

A "Corrected Amendment" filed under 37 C.F.R. 1.116, on March 23, 2007, has been entered pursuant to the Office Action of April 18, 2007.

**(v) SUMMARY OF CLAIMED SUBJECT MATTER**

The invention is directed to a catalyst composition (claims 1, 3-5 and 11-14), a method of producing the catalyst (claims 6-8), and a process for the selective hydrodesulphurization of feedstocks, comprising sulfur-containing compounds and olefins, employing the catalyst according to claim 1 (claims 9 and 10).

With respect to the catalyst composition, it requires a substrate which is a refractory oxide having a specific surface of at least  $150 \text{ m}^2/\text{g}$ , and at least one metal that is selected from groups VI and VIII of the periodic table, and carbon. The content of carbon which is a key to the invention. Claim 1 requires between 0.5 and 2.6% by weight of carbon, as supported by the specification on page 4, last complete paragraph. Likewise, claims 13 and 14 are directed to a carbon content between 1 and 2.6% by weight, again being supported by the last complete paragraph on page 4 of the specification.

The specific surface of less than  $150 \text{ m}^2/\text{g}$  is also important as when combined with the critical amount of carbon provides a marked improvement in the  $[K_{\text{HDS}}/K_{\text{HDO}}]$  selectivity as pointed out on page 5, fifth line, and the third complete paragraph. (The ratio is explained on page 3, second complete paragraph wherein  $(K_{\text{HDS}})$  is the rate (speed) constant to transform organic sulfur-containing compounds in the feedstock to  $\text{H}_2\text{S}$ , thereby permitting separation of same from the gasoline product, in order to comply with stricter standards for the maximum amount of sulfur that can be present in gasoline. Conversely, the rate constant  $K_{\text{(HDO)}}$  describes

the rate at which olefins in the feed are hydrogenated. Inasmuch as the olefins in the feed give rise to higher octane values, it is undesired to hydrogenate same. Consequently, the objective is to obtain a high  $K_{(HDS)}$  and a low  $K_{(HDO)}$  value.

Another aspect of the catalyst relates to the particular metals of group VI, molybdenum and tungsten and the metal of group VIII of nickel and cobalt, as set forth in claim 5 and more specifically in claim 11 which requires both cobalt and molybdenum and claim 12 which requires a catalyst comprising nickel oxide supported in (sic) alumina. The catalytic characteristics in performance levels of catalysts containing both CoO and MoO<sub>3</sub> is found in Table 1 on page 12 of the specification and Table 3 on page 14 of the specification, the latter table showing the importance of the specific surface area of the substrate, as well as Table 4 on page 15 of the specification. (These results are presented graphically in the argument section of this brief.)

**(vi) GROUNDS OF REJECTION TO BE REVIEWED ON APPEAL**

Referring to the FINAL REJECTION, claim 6 was rejected as being anticipated, but this claim was amended in the "Corrected Amendment under 37 C.F.R. 1.116) so that it is dependent on claim 1, so the patentability of claim 1 will inure to the patentability of claim 6 under the decisional law of *In Re Ochiai*. Accordingly, the following rejections are presented for review on appeal:

The rejection of claims 1, 2, 4, 5, 7-14 under 35 U.S.C. 103(a) as being unpatentable over Sadakane (EP 0745660) in view of Da Costa et al. U.S. 6,372,125. (These two references are described in Applicants' specification on page 2, last two paragraphs, but it is not apparent that they have been listed on PTO Form 892.)

Applicants also request review of the rejection of claim 3 as being unpatentable over Sadakane in view of Da Costa and further in view of Dufresne (U.S. 5,922,638) and also the rejection of claim 8 over Da Costa alone. Appellants will rely, however, on the patentability of claim 1 for supporting the patentability of claims 3 and 8 and will not argue such claims independently. (Note also that claim 3 is improperly dependent on cancelled claim 2 instead of claim 1. Authorization to make such a change is hereby granted.)

### **(vii) ARGUMENT**

Rejection of claims 1, 2, 4, 5, and 7-14 in the FINAL REJECTION as being unpatentable over Sadakane in view of Da Costa.

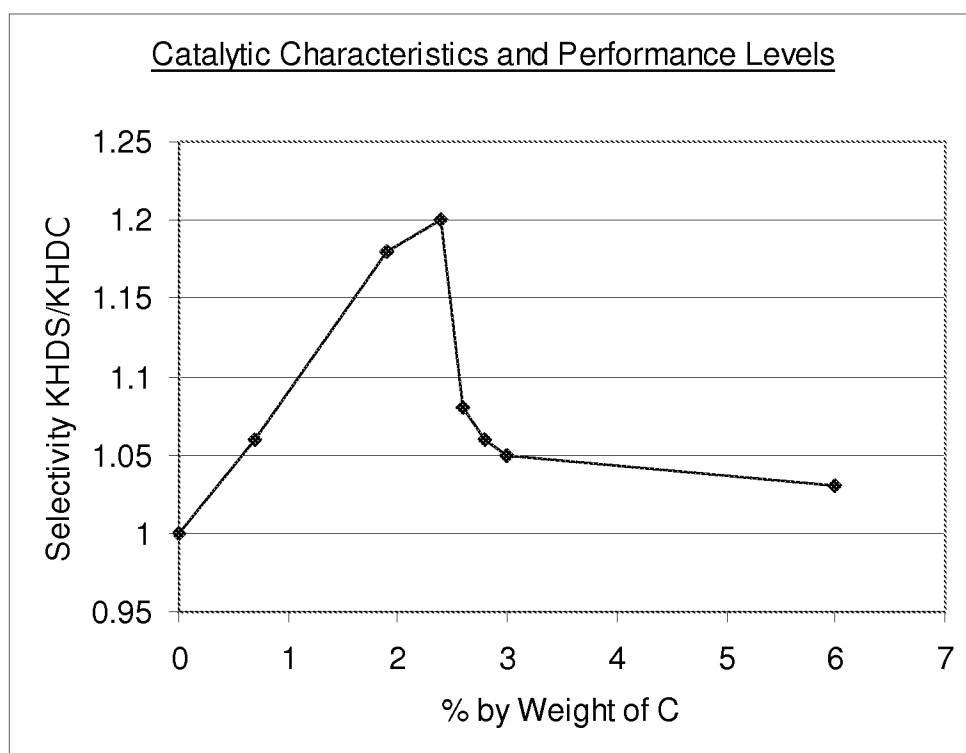
In the FINAL REJECTION, it is stated in paragraph 10 that Sadakane discloses that the catalyst has a carbon content of 2.1% (column 7, lines 31-34), the value of 2.1% being obtained by multiplying the 3.9% coke by the atomic ratio of carbon to hydrogen of 0.53.

The atomic ratio of carbon to hydrogen of 0.53 means that for 100 atoms of hydrogen there are 53 atoms of carbon. Consequently, since the atomic weight of carbon is 12 and the atomic weight of hydrogen is 1, it is then necessary to first obtain the weight ratio (the weight of carbon divided by the weight of carbon plus the weight of hydrogen, i.e.  $(53 \times 12)$  divided by  $100 + (53 \times 12)$  which equals 0.864. Thus, it is the value of 0.864 which is multiplied by 3.9% yielding 3.37%, not 2.1%. In Example 2, the content of carbon is much greater - 6.3%, in Example 3, 5.04%, and in Example 4, 7.73%. Thus, notwithstanding the statement on column 9, lines 37 and 38 that the coke deposited is in an amount of about 3 to about 10% by weight of the catalyst, there is no reason for one of ordinary skill in the art to employ a catalyst having an amount of carbon equal to between 0.5 and 2.6% by weight. If anything, the reference leads one of ordinary skill in the art away from such low contents of carbon deposits. Furthermore, Appellants obtain unexpected results by the utilization of a catalyst having a carbon content of between 0.5 and 2.6% by weight and a catalyst substrate having a specific surface of less than 150 m<sup>2</sup>/g.

The importance of the low concentration of carbon in the catalyst is demonstrated by Tables 1, 2 and 4. (Note that in the Tables the K values are described as activity constants, but they are described more correctly in the specification on page 3, second paragraph as reaction speed constants, normally termed in the U.S. as reaction rates. Nevertheless, for purposes of consistency, the K values in the Table are called activity constants in the following discussion.) Referring to Table 1 on page 12, the catalyst is supported by a substrate having a specific surface of 130 m<sup>2</sup>/g and wherein the catalyst has varying contents of carbon deposited thereon. Catalyst 4 containing 2.4% carbon has the highest activity constant for hydrodesulphurization and also the highest selectivity ratio of the rate of hydrodesulphurization versus hydrogenation of olefins. (It

will be recalled that the hydrogenation of olefins leads to a reduction in octane and is therefore not desired.) By examining the results of Table 1, one can note that the results based on carbon contents of 2.6 and below are higher than the results obtained with carbon contents above 2.6. Actually, as the carbon content is increased from 0.7 to 2.4, selectivity increases, and thereafter, and selectivity decreases, resulting in an unexpected discontinuity.

To facilitate comprehension of Table 1, the following is a graph which clearly shows Appellants' unexpected results achieved by employing a catalyst having an amount of carbon of between 0.5 and 2.6% by weight:



From the standpoint of case law, attention is invited to *In re Woodruff* (Fed. Cir.) 919 Fed. 2d 1575, 116 USPQ 2d 1934 which holds that where a range is disclosed in the prior art and the claimed range falls within that range but provides new and unexpected results relative to the prior art, the subject matter is unobvious.

Referring to Table 2, the results are even more pronounced since catalyst 10 having 0.9% carbon achieves an activity constant of 1.03 and selectivity of 1.28 compared to catalyst 11 having 5% by weight of carbon and showing an activity constant of 0.92 and selectivity of 1.10. Likewise, in the same manner, catalysts 15 and 16 achieve unexpected results, irrespective of the method of deposition since these catalysts contain 2.1% by weight of carbon.

With respect to the importance of the specific surface of the substrate, Table 3 on page 14 shows, all other variables being equal, that catalysts based on substrates having specific surfaces below 150 result in higher selectivities than catalysts having specific surfaces above 150. Referring to the Sadakane et al. reference, whereas there is no mention of specific surface therein, Applicants' French representative reports that supports for conventional hydrodesulphurization catalysts have high specific areas (typically higher than 200 m<sup>2</sup>/g).

Whereas the Da Costa reference discloses a support having a specific surface in the range of 100 to 600 m<sup>2</sup>/g (column 3, lines 26-23), this teaching would not lead one of ordinary skill in the art to Applicants combination of both a low carbon content of between 0.5 and 2.6% by weight and a low specific surface area of less than 150 m<sup>2</sup>/g, much less the unexpected and highly advantageous results that are achieved by the utilization of such catalysts.

In view of the above discussion, it is respectfully submitted that claim 1, describing a catalyst, which achieves unexpected results, is truly unobvious and patentable under 35 U.S.C. 103 in this highly crowded art of the hydrodesulphurization of hydrocarbon feedstocks.

Referring now to the dependent claims, it is seen that claim 4 restricts the specific surface area to not more than 130 m<sup>2</sup>/g.

At this juncture, attention is courteously invited to section 2144.08, page 2100-160 in the recent Manual of Patenting Examining Procedure revision of August 2005 wherein the left hand column, last paragraph states that a showing of unexpected results for a single member of a claimed subgenus, or a narrow portion of a claimed range would be sufficient to rebut a prima facie case of obviousness if a skilled artisan could ascertain a trend in the exemplified data that would allow him to reasonably extend the probative value thereof. *In re Clemens* 62 Fed. 2nd 1029, 1036 206 USPQ 289, 296 (CCPA 1980), (evidence of the unobviousness of a broad range can be proven by a narrower range when one skilled in the art could ascertain the trend that

would allow him to reasonably extend the probative value thereof). It is respectfully submitted that the above discussion of the unexpected results shown in the Tables can reasonably be extended to Applicants' ranges in the claims.

Accordingly, Appellants respectfully urge that the Examiner's rejection be reversed.

The Commissioner is hereby authorized to charge any fees associated with this response or credit any overpayment to Deposit Account No. 13-3402.

Respectfully submitted,

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**(viii) CLAIMS APPENDIX**

Claim 1. A catalyst for selective hydrodesulfurization of hydrocarbon feedstocks that comprise sulfur-containing compounds and olefins, said catalyst comprising a substrate that is selected from among the refractory oxides having a specific surface of less than  $150 \text{ m}^2/\text{g}$ , at least one metal that is selected from the group that consists of the metals of groups VI and VIII of the periodic table and carbon, characterized in that the carbon content is between 0.5 and 2.6% by weight and in that the catalyst is in a sulfide form.

Claim 2. Cancelled.

Claim 3. A catalyst according to claim 2, wherein the overall sulfur content in said catalyst is between 60 and 140% of the sulfur content that is necessary for the total sulfurization of all of the metals of said catalyst belonging to groups VI and VIII.

Claim 4. A catalyst according to claim 1, wherein the specific surface area of the substrate of not more than  $130 \text{ m}^2/\text{g}$ .

Claim 5. A catalyst according to claim 1, wherein the metal of group VI is selected from the group that consists of molybdenum and tungsten, and the metal of group VIII is selected from the group that consists of nickel and cobalt.

Claim 6. A method for the production of a catalyst according to claim 1 for selective hydrodesulfurization of hydrocarbon-containing feedstocks that comprise sulfur-containing compounds and olefins, said method comprising:

- a stage for impregnation of metals of groups VI and/or VIII on a substrate,
- an activation stage that is a sulfurization stage that is carried out by



- contact with a gas that comprises hydrogen and hydrogen sulfide, and
- a stage for deposition of carbon by contact with at least one hydrocarbon-containing compound, so as to deposit an amount of carbon that is between 0.5 and 2.6% by weight relative to the mass of catalyst.

Claim 7. A method according to claim 6, wherein the stage for deposition of carbon is carried out during the activation stage.

Claim 8. A method according to claim 6, wherein the stage for deposition of carbon is carried out at the same time as the impregnation of metals of groups VI and/or VIII by depositing a precursor that contains carbon at the time of impregnation of the metals of groups VI and/or VIII.

Claim 9. A process for the selective hydrodesulfurization of feedstocks that comprise sulfur-containing compounds and olefins, wherein said process comprises using the catalyst according to claim 1.

Claim 10. A process according to claim 9, wherein the feedstock comprises a gasoline fraction that is obtained from a catalytic cracking unit containing hydrocarbons with 5 carbon atoms to compounds that have a boiling point of approximately 250°C.

Claim 11. A catalyst according to claim 1, comprising cobalt and molybdenum.

Claim 12. A catalyst according to claim 1, comprising nickel oxide supported in alumina.

Claim 13. A catalyst according to claim 1, having a carbon content of 1 to 2.6% by weight.

Claim 14. A catalyst according to claim 11, having a carbon content of 1 to 2.6% by weight.

**(ix) EVIDENCE APPENDIX**

{ none }

**(x) RELATED PROCEEDINGS APPENDIX**

{ none }